

# Non-equilibrium growth in a restricted-curvature model

Amit Kr. Chattopadhyay

*Dept. of Theoretical Physics,*

*Indian Association for the Cultivation of Science,*

*Jadavpur, Calcutta 700 032*

The static and dynamic roughenings of a growing crystalline facet is studied where the growth mechanism is controlled by a restricted-curvature (RC) geometry. A continuum equation, in analogy with the Kardar-Parisi-Zhang (KPZ) equation is considered for the purpose. It is shown here that although the growth process begins with a RC geometry, a structural phase transition occurs from the restricted-curvature phase to the KPZ phase. An estimation of the corresponding critical temperature is given here. Calculations on the static phase transition give results along the same line as existing predictions, apart from minor numerical adjustments.

PACS number(s): 05.40.+j, 68.35.Rh, 64.40.Ht

Growth of interfaces in a strongly temperature controlled regime has been the subject of a considerable portion of recent technological developments [1]. The interest in this field has mainly been generated by the microscopic roughness that originates as a result of competition among different effects, such as surface tension, thermal diffusion and different noise factors coming into play during the growth process. Even in equilibrium, a crystalline facet "remains practically flat until a transition temperature is reached, at which the roughness of the surface increases very rapidly" [2]. Numerous theoretical models, starting from Burton, et al [2] have been proposed to account for a detailed analysis of the height fluctuations during the growth process on both sides of the roughening temperature  $T_R$  [3-5]. They found a change in mobility from activated growth in the nucleated phase (characterized by low temperatures) to nonactivated growth at large temperatures. The pinning-depinning growth model of Chui and Weeks [3] has later been numerically extended to a polynuclear growth model by Sarloos and Gilmer [6] confirming a growth by island formation which is continuously destroyed by any small chemical potential favoring the growth. Theoretical forays in this front have continued with detailed predictions on the equilibrium roughening transition [7,8] which have also found experimental justifications in [9].

Starting from the early theoretical efforts [2,3] to the present day developments [1,10], numerous discrete [11,12] and continuum [13,14] models have been proposed to study both equilibrium and nonequilibrium surface properties. However all these different models seem to fall within either of the KPZ or the Lai-Das Sarma universality class, barring a few exceptions [11,15]. The atomistic growth process of the latter type deals with surfaces grown by molecular beam epitaxy (MBE) method, whose distinguishing feature is that growth occurs under surface diffusion conditions, with the deposited atoms relaxing to the nearby kinks. The essential idea employed was to modify the relaxation mechanism as a locally sur-

face minimizing curvature, instead of surface area. To linear order, this mechanism was supposed to mimic the growth dynamics of crystalline surfaces [12,14].

Alternative efforts in this front have mainly centered around the development of theoretical models whose dynamics can be mapped to either of these two main universality classes. A classic example is an equilibrium restricted-curvature (RC) model studied by Kim and Das Sarma [16]. The corresponding growth rule describes a growth restricted on the local curvature,  $|\nabla^2 h| \leq N$ , and is obeyed at both the growing site and its nearest neighbors where  $N$  is any fixed positive integer. We adopt the nonequilibrium class of growth proposed by Kim and Das Sarma as the starting point of our study of the dynamics of the nonequilibrium growth in a MBE process.

Considering two dimensional growth pertaining to a lattice structure where  $h(\vec{r}, t)$  is the height of the interface at time  $t$  at position  $\vec{r}$ , the equation goes like

$$\eta \frac{\partial h}{\partial t} = -\gamma \nabla^4 h(\vec{r}, t) + \frac{\lambda}{2} |\nabla^2 h(\vec{r}, t)|^2 - \frac{2\pi V}{a} \sin\left[\frac{2\pi}{h}(\vec{r}, t)\right] + F + R(\vec{r}, t) \quad (1)$$

where  $\eta$  is the inverse mobility which fixes the time scale,  $\gamma$  is the surface tension,  $a$  is the lattice constant and  $V$  is the strength of the pinning potential.  $F$  is a steady driving force with  $R$  the white noise defined as

$$< R(\vec{r}, t) R(\vec{r}', t') > = 2D\delta^2(\vec{r} - \vec{r}')\delta(t - t') \quad (2)$$

The biharmonic term on the right side of the above equation gives the basic relaxation mechanism in operation. The second term is the most important term for the restricted-curvature (RC) dynamics and has been incorporated in analogy with the KPZ equation [13]. Just as the lateral term  $|\vec{\nabla} h|^2$  gives the nonlinearity in a KPZ growth, the curvature dependence in our model is proposed to produce a leading order nonlinearity of the form

$(\nabla^2 h)^2$ . From phenomenological considerations, since the discretisation scheme prohibits a high curvature, the constant  $\lambda$  is negative. The third term comes along due to the lattice structure preferring integral multiples of  $h$ , which means that  $h$  is measured in units of  $a$ .  $F$  is the steady driving force required to depin a pinned interface while all the microscopic fluctuations in the growth process are assimilated in the noise term  $R$ . It can be shown easily that the second and third terms are obtained from a variant of the sine-Gordon Hamiltonian

$$E[h(\vec{r}, t)] = \int \int d^2 r \left[ \frac{\gamma}{2} (\nabla^2 h)^2 - V \cos\left(\frac{2\pi}{a} h\right) \right] \quad (3)$$

We now start pursuing the most important goal regarding the dynamics of the growing interface - whether a non-equilibrium phase transition exists or not. In other words, whether we can specify a critical temperature  $T_R$  below which the surface is flat and above which the surface starts showing a dynamic roughening. This obviously brings into question the interplay of strengths between the non-linear term  $(\nabla^2 h)^2$  and the sine-Gordon potential. Three limits of the above eqn.(1) are well known:

- (i)  $\lambda = V = F = 0$ ; characterizing a stationery interface [11,12,16]. This sort of growth has found experimental justification in the works of Yang, et al and Jeffries, et al [19].
- (ii)  $\lambda = F = 0$ ,  $V \neq 0$ ; characterizing the growth dynamics of crystalline tensionless surfaces [20]. This model in equilibrium depicts a roughening transition to the high temperature regime of the sine-Gordon model and is expected to modelise the vacuum vapor deposition dynamics by MBE growth [21].
- (iii)  $V = F = 0$ ,  $\lambda \neq 0$ ; a very special case of the numerical simulation predicting a "local model" for dendritic growth with analytics failing to define a stable, non-trivial fixed point [22]. This issue is still under much debate and really needs a deeper understanding to have any final say in the matter.

However in both the first and second situations, detailed numerical analysis have predicted a transition from the conserved MBE growth to the Edwards- Wilkinson [23] type phase characterized by the generation of a  $\nabla^2 h$  term [20,24]. The generation of this so-called "surface tension" term in the dynamics although surely being a fall-out of the renormalization of the sine-Gordon potential in the latter model, actually demands a greater attention. Combining this idea with the proposition put forward by Kim and Das Sarma [16], we therefore pose the most general situation concerning a surface growing under MBE and try to ascertain the associated roughening process throughout the whole temperature range, with the growth essentially occuring under a surface curvature constraint. In the following analysis, we employ standard dynamic renormalization techniques to probe

the dynamics of our proposed Langevin equation (1) [7,8] and later on follow the line of Chui and Weeks [3,7] in taking account of the static renormalization of the model Hamiltonian shown in eqn.(3).

As usual, we start by first integrating over the momentum shell  $\Lambda(1 - dl) < |\vec{k}| < \Lambda$  perturbatively in  $\lambda$  and  $V$ . Thereafter we rescale back the variables in the form  $\vec{k} \rightarrow \vec{k}' = (1 + dl)\vec{k}$ ,  $h \rightarrow h' = h$  and  $t \rightarrow t' = (1 - 4dl)t$ . The various coefficients follow the rescaling  $\eta \rightarrow \eta' = \eta$ ,  $\gamma \rightarrow \gamma' = \gamma$ ,  $\lambda \rightarrow \lambda' = \lambda$ ,  $V \rightarrow V' = (1 + 4dl)V$ ,  $F \rightarrow F' = (1 + 4dl)F$ . We set up the perturbative scheme to rewrite eqn.(2) in a comoving frame moving with velocity  $F/\eta$  as

$$\eta \frac{\partial}{\partial t} h = -\gamma \nabla^4 h + \Phi(h) + R \quad (4)$$

where  $\Phi(h) = -\frac{2\pi V}{a} \sin[\frac{2\pi}{a}(h + \frac{F}{\eta}t)] + \frac{\lambda}{2} (\nabla^2 h)^2$ . Thereafter going exactly by the analysis of Nozieres and Gallet and Rost and Spohn [7,8] and including the corrections in [25], we finally arrive at an expression for the renormalized mode coupling term,

$$\begin{aligned} \Phi^{SG} = & -\frac{2\pi^3 V^2 T}{\gamma^2 a^5} dl \int_{-\infty}^t \int d^2 r' \frac{1}{t - t'} J_0(\Lambda(|\vec{r} - \vec{r}'|)) \\ & G_0(|\vec{r} - \vec{r}'|, t - t') \times e^{-[\frac{\gamma}{\eta} \Lambda^4 (t - t') + \frac{2\pi T}{a^2 \gamma} \phi(|\vec{r} - \vec{r}'|, t - t')]} \\ & \times [\frac{2\pi}{a} [\frac{\partial}{\partial t} \bar{h}(\vec{r}, t)(t - t') - \frac{1}{2} \partial_i \partial_j \bar{h}(\vec{r}, t)(r_i - r'_j) \times \\ & (r_j - r'_j)] \cos[\frac{2\pi F}{a \eta}(t - t')] + [1 - \frac{2\pi^2}{a^2} [\partial_i \bar{h}(\vec{r}, t)]^2 \\ & \times (r_i - r'_i)^2] \sin[\frac{2\pi F}{a \eta}(t - t')]] \end{aligned} \quad (5)$$

where  $\Lambda \sim 1/a$  is a suitably chosen upper cut-off with the Green's function  $G(x, t)$  given by

$$G(x, t) = \int dk e^{ikx - \nu t k^4} \quad (6)$$

and

$$\phi(\tilde{\rho}, x) = \int_0^\Lambda \frac{dk}{k} [1 - J_0(k\tilde{\rho})] e^{-\frac{\gamma}{\eta} k^2 (t - t')} \quad (7)$$

with  $\tilde{\rho} = \Lambda \rho$  and  $x = \frac{\gamma(t - t')}{\eta \rho^2}$ .

Turning now to the above eqn.(5), we find that starting with a structurally non-linear term in the Langevin equation, the dynamic renormalization has initiated a mode coupling structure with quite a different composition. The absence of the nonlinear  $(\nabla^2 h)^2$  term in  $\Phi^{SG}$  implies that once starting with a restricted curvature model of growth, The lattice structure partakes a dynamics where in a finite time after the start,  $\lambda$  is renormalized to zero and thereafter the KPZ type [13] nonlinearity takes over. Thus a competition ensues between the alternate pinning

and depinning forces offered by the  $(\nabla h)^2$  and  $(\nabla^2 h)^2$  nonlinearities. Also the production of the surface tension term  $\nabla^2 h$  ensures a dynamic phase transition from the restricted-curvature regime to the KPZ regime. From an analysis of the following renormalization flows, we arrive at an expression for the temperature at which the transition occurs:

$$\frac{dU}{dl} = (4 - n)U \quad (8)$$

$$\frac{d\gamma'}{dl} = \frac{2\pi^4}{\gamma a^4} nA^{(\gamma')}(n; \kappa) U^2 \quad (9)$$

$$\frac{d\gamma}{dl} = \frac{\pi^4}{6\gamma a^4} nA^{(\gamma)}(n; \kappa) U^2 \quad (10)$$

$$\frac{d\eta}{dl} = \frac{8\pi^4}{\gamma a^4} \frac{\eta}{\gamma} nA^{(\eta)}(n; \kappa) U^2 \quad (11)$$

$$\frac{d\lambda}{dl} = 0 \quad (12)$$

$$\frac{d\lambda'}{dl} = \frac{8\pi^5}{\gamma a^5} nA^{(\lambda')}(n; \kappa) U^2 \quad (13)$$

$$\frac{dK}{dl} = 4K + \frac{D}{4\pi\eta\gamma} \lambda - \frac{4\pi^3}{\gamma a^3} nA^{(K)}(n; \kappa) U^2 \quad (14)$$

$$\frac{dD}{dl} = \frac{1}{8\pi} D \frac{\lambda^2 D}{\gamma^3} + \frac{8\pi^4}{\gamma a^4} \frac{D}{\gamma} nA^{(n)}(n; \kappa) U^2 \quad (15)$$

where  $U = \frac{V}{\Lambda^2}$ ,  $K = \frac{F}{\Lambda^2}$ ,  $\kappa = \frac{2\pi K}{a\gamma}$  and  $n = \frac{\pi T}{\gamma a^2}$ , with  $\gamma'$  representing the coefficient of the renormalized surface term [26] and  $\lambda'$  denoting the coefficient corresponding to the KPZ nonlinearity generated on account of renormalization.  $A^{(i)}(n; \kappa)$  stand as shorthand representation for the integrals, where  $i = \eta, \lambda', \lambda, \kappa$ , etc. and whose detailed functional forms are given below:

$$A^{(\gamma')}(n; \kappa) = \int_0^\infty \frac{dx}{x} \int_0^\infty d\tilde{\rho} \tilde{\rho}^3 J_0(\tilde{\rho}) \times \cos\left[\frac{2\pi}{a} \frac{Kx\tilde{\rho}^2}{\gamma}\right] \times \int dp e^{\frac{\tilde{\kappa}}{\lambda}(ip - \frac{\kappa}{\lambda} \frac{2}{\gamma} \tilde{\rho} x p^4)} \times e^{-[x\tilde{\rho}^2 + 2n\phi(\tilde{\rho}, x)]} \quad (16)$$

$$A^{(\eta)}(n; \kappa) = \int_0^\infty dx \int_0^\infty d\tilde{\rho} \tilde{\rho} J_0(\tilde{\rho}) \times \cos\left[\frac{2\pi}{a} \frac{Kx\tilde{\rho}^2}{\gamma}\right] \times \int dp e^{\frac{\tilde{\kappa}}{\lambda}(ip - \frac{\kappa}{\lambda} \frac{2}{\gamma} \tilde{\rho} x p^4)} \times e^{-[x\tilde{\rho}^2 + 2n\phi(\tilde{\rho}, x)]} \quad (17)$$

$$A^{(\lambda')}(n; \kappa) = \int_0^\infty \frac{dx}{x} \int_0^\infty d\tilde{\rho} \tilde{\rho}^3 J_0(\tilde{\rho}) \times \sin\left[\frac{2\pi}{a} \frac{Kx\tilde{\rho}^2}{\gamma}\right] \times \int dp e^{\frac{\tilde{\kappa}}{\lambda}(ip - \frac{\kappa}{\lambda} \frac{2}{\gamma} \tilde{\rho} x p^4)} \times e^{-[x\tilde{\rho}^2 + 2n\phi(\tilde{\rho}, x)]} \quad (18)$$

$$A^{(K)}(n; \kappa) = \int_0^\infty \frac{dx}{x} \int_0^\infty d\tilde{\rho} \tilde{\rho} J_0(\tilde{\rho}) \times \sin\left[\frac{2\pi}{a} \frac{Kx\tilde{\rho}^2}{\gamma}\right] \times \int dp e^{\frac{\tilde{\kappa}}{\lambda}(ip - \frac{\kappa}{\lambda} \frac{2}{\gamma} \tilde{\rho} x p^4)} \times e^{-[x\tilde{\rho}^2 + 2n\phi(\tilde{\rho}, x)]} \quad (19)$$

$$A^{(\gamma)}(n; \kappa) = \int_0^\infty \frac{dx}{x} \int_0^\infty d\tilde{\rho} \tilde{\rho}^5 J_0(\tilde{\rho}) \times \cos\left[\frac{2\pi}{a} \frac{Kx\tilde{\rho}^2}{\gamma}\right] \times \int dp e^{\frac{\tilde{\kappa}}{\lambda}(ip - \frac{\kappa}{\lambda} \frac{2}{\gamma} \tilde{\rho} x p^4)} \times e^{-[x\tilde{\rho}^2 + 2n\phi(\tilde{\rho}, x)]} \quad (20)$$

As already argued, the eqn.(13) giving the flow for the renormalized KPZ coefficient generates the lattice potential of the driven interface after the structural phase transition has occurred. At some temperature  $T_R = \frac{4\gamma a^2}{\pi}$ , in the units measured, corresponding to the transition point  $n = 4$ , the roughness dynamics driven by the KPZ force takes over. From this point the whole dynamics follows in the line predicted by Nozieres and Gallet [7] and Rost and Spohn [8], with the biharmonic and structural nonlinear terms muffled by the surface tension term and the KPZ nonlinearity respectively. This KPZ regime continues until the growing interface reaches the next crystalline facet whereon the RC dynamics again comes into play and the whole mechanism goes on repeating itself throughout the process of nonequilibrium growth. However in the static case, starting with the standard sine-Gordon Hamiltonian (eqn.(3)) and again taking clues from [7,8], we arrive at the following Kosterlitz-Thouless type second order equations [5,10] in terms of the reduced variables  $y = \frac{4\pi U}{T}$  and  $x = \frac{4\gamma a^2}{\pi T}$ ,

$$\frac{dx}{dl} = \frac{y^2}{x} B(4/x) \quad (21)$$

$$\frac{dy}{dl} = 4y(1 - \frac{1}{x}) \quad (22)$$

where

$$B(n) = \int_0^\infty d\tilde{\rho} \tilde{\rho}^3 J_0(\tilde{\rho}) e^{-2nh(\tilde{\rho})} \quad (23)$$

and

$$h(\tilde{\rho}) = \int_0^\Lambda \frac{dk}{k^3} [1 - J_0(k\tilde{\rho})] \quad (24)$$

with  $n = \frac{\pi T}{\gamma a^2}$ . The roughening transition occurs at the fixed point  $y = 0$ ,  $x = 1$  and we recover all the predictions of Nozieres, [10] although with slightly different

coefficients. The point to be noted is that although the transition temperature is twice here compared to that of the standard Kosterlitz-Thouless (KT) case, both the dynamic and static phase transitions occur at the same critical temperature  $T_R = \frac{4\gamma a^2}{\pi}$ , somewhat alike to the NG and KT models.

In conclusion, we have studied interface growth models both in the equilibrium and nonequilibrium cases under a constraint of the growth occurring under a curvature restriction. The dynamic model gives a phase transition from the RC growth to the KPZ type at a temperature we have calculated to be  $\frac{4\gamma a^2}{\pi}$ . Experimental data on the roughening transition of solid  $^4\text{He}$  in contact with the superfluid  $^3\text{He}$  [9] seems to show a crossover to the non-equilibrium regime in the superfluid phase. We suspect that in a scenario analogous to the MBE growth model proposed, the crossover situation is actually mimicked by the dynamic phase transition found in the model discussed and the larger value of the critical phase transition temperature reported here might just be the explanation for the weaker coupling observed around  $0.1^\circ\text{K}$  in  $^3\text{He}$ . However, although the dynamic mechanism proposed here differs largely from the other existing models, thereby defining a new universality class, the behavior at equilibrium defined by the corresponding static model is actually a replica of the Kosterlitz-Thouless criticality, the only variation being in an augmented value of the transition temperature.

The author acknowledges partial financial support from the Council of Scientific and Industrial Research, India during the whole tenure of this work. Also sincere gratitude goes to Prof. Jayanta K. Bhattacharjee for numerous illuminating discussions with him during the course of this work.

- S. Balibar, E. Rolley and P. Nozieres, J. Phys. (Paris) **46**, 1987 (1985).
- [10] J. Krug and H. Spohn; P. Nozieres in "Solids Far From Equilibrium" edtd. by C. Godreche, Cambridge Univ. Press, NY, 1992.
  - [11] S. Das Sarma and P. I. Tamborenea, Phys. Rev. Letts, **66**, 325 (1991).
  - [12] P. E. Wolf and J. Villain, Euro Phys. Letts. **13**, 389 (1990); F. Family and T. Vicsek, J. Phys. A **18**, L75 (1985).
  - [13] M. Kardar, G. Parisi and Y. C. Zhang, Phys. Rev. Letts. **56**, 889 (1986).
  - [14] Z. W. Lai and S. Das Sarma, Phys. Rev. Letts. **66**, 2348 (1991).
  - [15] J. Krug, Adv. in Phys. **46**, 139 (1997).
  - [16] J. M. Kim and S. Das Sarma, Phys. Rev. E **48**, 2599 (1993).
  - [17] J. Villain, J. Phys. (Paris) I **1**, 19 (1991).
  - [18] S. Das Sarma and S. V. Ghaisas, Phys. Rev. Letts. **69**, 3758 (1992).
  - [19] H. N. Yang, Y. P. Zhao, G. C. Wang and T. M. Lu, Phys. Rev. Letts. **76**, 3774 (1996); J. H. Jeffries, J. F. Zuo and M. M. Craig, Phys. Rev. Letts. **76**, 4931 (1996).
  - [20] E. Moro, R. Cuerno and A. Sanchez, Phys. Rev. Letts. **78**, 4982 (1997).
  - [21] A. Zhangwill, J. Cryst. Growth **163**, 8 (1996).
  - [22] N. Govind and H. Guo, J. Phys. **25**, 5485(1992).
  - [23] S. F. Edwards, D. R. Wilkison, Proc. R. Soc. A **381**, 17 (1982).
  - [24] J. G. Amar, P. M. Lam and F. Family, Phys. Rev. E **47**, 3242 (1993).
  - [25] H. J. F. Knops and L. W. Den Ouden, Physica A **103**, 579 (1980).
  - [26] S. Das Sarma and R. Kotlyar, Phys. Rev. E **50**, R4275 (1994).

- 
- [1] A. L. Barabasi and E. Stanley, "Fractal Concepts in Surface Growth", Cambridge Univ. Press, NY, 1995.
  - [2] W. K. Burton, N. Cabrera and F. C. Frank, Philos. Trans. R. Soc. **243A**, 299 (1951).
  - [3] S. T. Chui and J. D. Weeks, Phys. Rev. B **14**, 4978 (1976).
  - [4] H. van Beijeren, Phys. Rev. Letts. **38**, 993 (1977).
  - [5] J. M. Kosterlitz and D. J. Thouless, J. Phys. C **6**, 1181 (1973); J. Phys. C **7**, 1046 (1974).
  - [6] W. van Sarloos and G. H. Gilmer, Phys. Rev. B **33**, 4927 (1986).
  - [7] P. Nozieres and F. Gallet, J. Phys. (Paris) **48**, 353 (1987).
  - [8] M. Rost and H. Spohn, Phys. Rev. E **49**, 3709 (1991).
  - [9] F. Gallet, S. Balibar and E. Rolley, J. Phys. (Paris) **48**, 369 (1987); Y. Carmi, E. Polturak and S. G. Lipson, Phys. Rev. Letts, **62**, 1364 (1989); P. E. Wolf, F. Gallet,